A Study of Separation Efficiency in Thermal Diffusion Columns with a Permeable Vertical Barrier

A separation theory in thermal diffusion columns with a permeable vertical barrier has been derived by making use of the orthogonal expansion method. It is shown that the undesirable remixing effect can be effectively reduced and controlled by introducing a permeable vertical barrier. Considerable improvement in separation was obtained when the position of the barrier was carefully chosen.

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SCOPE

Thermal diffusion takes place when a temperature gradient in a mixture of two gases or liquids gives rise to a concentration gradient with one component concentrated near the hot wall and the other component concentrated near the cold wall. This process can be used to separate mixtures that are hard to separate by conventional methods such as distillation and extraction. It was the great achievement of Clusius and Dickel (1938) to introduce the thermogravitational thermal diffusion column to make this separation process practical. From then on, many improved columns, such as packed, inclined, wired, rotary, inclined moving-wall, and rotated wired columns, have been proposed to improve the separation of the thermogravitational thermal diffusion column. Treacy and Rich (1955) have found that installing a permeable vertical barrier in the working space of the column can also effectively increase both the magnitude and the rate of separation. Basically, the transport phenomena in this column belong to the category of conjugated Graetz problems with recycle at both ends. Difficulties in such problems occur because neither the interfacial concentration nor the concentrations at the ends can be specified a priori. It is the purpose of this work to develop a separation theory of this column thoroughly by use of an orthogonal expansion technique. The results obtainable in this work may also be used with other conjugated Graetz problems with countercurrent flow and with reflux internally or externally at both ends.

CONCLUSIONS AND SIGNIFICANCE

The equation of separation in a thermal diffusion column with a permeable vertical barrier has been derived by using the orthogonal expansion method. Some graphical solutions of the degree of separation obtained from theoretical developments were compared with those obtained from experiments at various flow rates and barrier positions. With careful choice of the barrier position, considerable improvement in separation is reached compared to the Clusius-Dickel columns.

Introduction

The thermogravitational thermal diffusion column, introduced by Clusius and Dickel (1938), is an unusual device for the separation of liquid or gas mixtures that are difficult or impossible to separate by ordinary means. It was used to separate uranium isotopes at Oak Ridge Laboratory in World War II, and a commercial plant to produce lubricating oil with a high viscosity index has also been designed. The Clusius-Dickel column consists essentially of two opposing vertical plates separated by a very narrow open space. One plate is heated and the other cooled, and the thermal diffusion effect causes one component of, say, a binary mixture to diffuse toward the hot plate. At the same time the density gradient that arises because of the temperature gradient causes smooth laminar convection currents up the hot plate and down the cold plate. Because of the concentration gradient set up by the thermal diffusion, the convection currents transport one component preferentially toward the top and thus create large concentration differences between the top and the bottom of the column. The first complete presentation of the theory of the Clusius-Dickel column was made by Furry et al. (1939). A more detailed study of the mechanism of separation in the Clusius-Dickel column indicates that the convective currents actually have two conflicting effects: a desirable cascading effect and an undesirable remixing effect. At steady state, a dynamic equilibrium is established between these two effects and hence no further separation takes place. It is thus evident that any improvement in the equilibrium separation must be associated with either a suppression of the remixing effect and/or an enhancement of the cascading effect. It is possible, therefore, that proper adjustment of the convective strengths might effectively improve the separation. Based on this concept, some improved columns have been developed in the literature, such as in the cases of packed (Lorenz and Emery, 1959; Yeh and Chu, 1974), inclined (Powers and Wilke 1957, Cheuh and Yeh 1967), wired (Washall and Molpolder, 1962; Yeh and Ward, 1971), rotary (Sullivan et al., 1955; Yeh and Cheng, 1973; Yeh and Tsai, 1981, 1982), and inclined moving-wall and rotated wired columns (Ramser, 1957; Yeh and Tsai, 1972; Yeh and Ho, 1975; Yeh and Tsai 1981).

In 1955, Treacy and Rich examined experimentally the effect of installing several types of barrier in the working space of the column. According to them, the introduction of thin permeable verticle barriers resulted in increases of both the magnitude and the rate of separation. Later, Bobrova and Rabinovich (1971), Sasaki et al. (1976, 1977) and Selecki et al. (1980) derived the separation equation of a barrier column based on the theory suggested by Furry et al. (1939). Yet, in order to match the experimental data, Babrova and Rabinovich and Selecki et al. have introduced a parameter during the derivation of the column theory. Even so, the result obtained by Selecki et al. was still qualitatively in agreement with the experimental data.

Since the transport phenomena in this kind of column belongs to the category of conjugated boundary value problems (Nunge and Gill, 1966; Davis, 1973; Davis and Venkatesh, 1979; Papoutsakis and Ramkrishna 1981a, b), there is no doubt that analysis based on the theory proposed by Furry et al. will fail. The present work actually deals with extended conjugated Graetz problems with recycle at both ends. The purpose of this paper is to report on an experimental study, and thus to complete the theoretical development by use of orthogonal expansion techniques concerned with eigenfunctions (Churchill and Brown, 1978).

Theoretical Development

Consider a flat-plate thermal diffusion column with a permeable vertical barrier inserted between the plates, as shown in Figure 1. The barrier has a thickness comparable with the distance between the plates. Feeds are introduced from the center of the column and the products are withdrawn from both ends continuously with equal flow rates. The whole column is composed of two sections and each section is divided into two regions by the barrier. Fully developed fluids in both regions flow countercurrently with internal reflux at the ends. The coordinates are also shown in Figure 1 with origins located at the feed points.

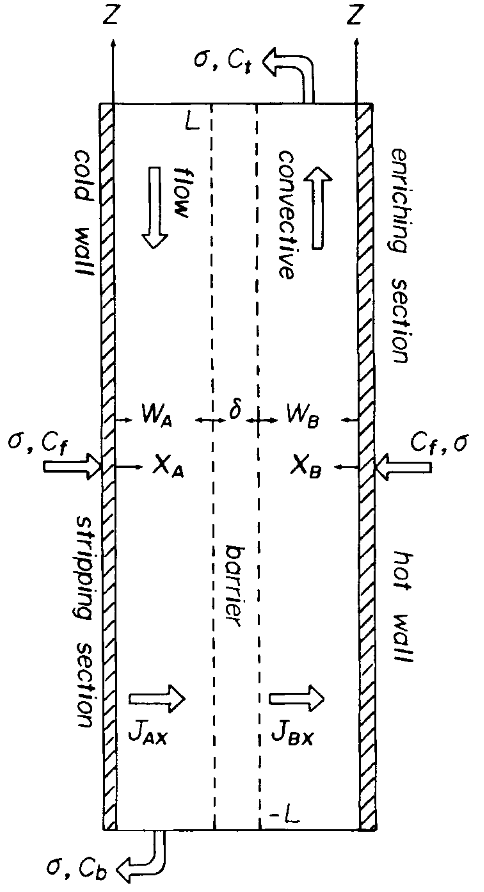

Figure 1. Continuous-flow thermal diffusion column with vertical barrier.

Temperature distributions

Since the space between the plates is small, the heat transfer is determined only by conduction. Accordingly, the temperature distribution in the fluid is linear and the temperature gradient of the fluid in the regions is

$$h_1 = \frac{\Delta T}{W_A + W_B + \dfrac{K_0\delta}{K_0\epsilon + K(1-\epsilon)}} \tag{1}$$

where K_0 is the thermal conductivity of the fluid, and K, δ, and ϵ are the thermal conductivity, thickness, and permeability of the barrier, respectively. During the derivation of Eq. 1 we assumed that the thermal resistance in the barrier was composed of the resistances of the fluid and the barrier in parallel. Hence, the temperature gradient of the fluid within the barrier can be expressed as

$$h_2 = \frac{h_1 K_0}{K_0\epsilon + K(1-\epsilon)} \tag{2}$$

Velocity distributions

At steady state, the velocity distribution in axial direction is caused by:

1. Natural convection
2. Additional convective strengths due to the difference between the mean temperatures of fluids in both regions.
3. Forced convection.

Assuming laminar flows of the fluid in the regions and neglecting the influences of ordinary and thermal diffusions, end effects, and inertia terms on the velocity, we obtain the momentum equations in the enriching section as follows:

$$\frac{d^2V_{Ae}(\eta_A)}{d\eta_A^2} = \frac{-\beta g W_A^2}{\mu}(T_1 - \tilde{T} + h_1 W_A \eta_A) + \frac{W_A^2}{\mu}\left(\frac{dP}{dz}\right) \quad (3)$$

$$\frac{d^2V_{Be}(\eta_B)}{d\eta_B^2} = \frac{-\beta g W_B^2}{\mu}(T_2 - \tilde{T} - h_1 W_B \eta_B) + \frac{W_B^2}{\mu}\left(\frac{dP}{dz}\right) \quad (4)$$

where the pressure drop dP/dz is caused by forced convection and the dimensionless coordinates are defined in the Notation. With the nonslip conditions at the boundaries of the walls and the barrier, and Eq. 5 obtained from material balance, we may obtain the velocity profiles in the enriching section. The reference temperature $\tilde{T}$ is determined by Eq. 5 with σ set to zero (no feed introduced):

$$\rho B\left[W_A \int_0^1 V_{Ae}(\eta_A)d\eta_A + W_B \int_0^1 V_{Be}(\eta_B)d\eta_B\right] = \sigma \quad (5)$$

Hence, the resulting equations are

$$\tilde{T} = \frac{2W_A^3 T_1 + 2W_B^3 T_2 - h_1(W_B^4 - W_A^4)}{2(W_A^3 + W_B^3)} \quad (6)$$

$$V_{Ae}(\eta_A) = f_{1e}(\eta_A - \eta_A^3) + f_{2e}(\eta_A^2 - \eta_A) + f_{3e}(\eta_A - \eta_A^2) \quad (7)$$

$$V_{Be}(\eta_B) = g_{1e}(\eta_B^3 - \eta_B) + g_{2e}(\eta_B - \eta_B^2) + g_{3e}(\eta_B - \eta_B^2) \quad (8)$$

in which

$$f_{1e} = \frac{\beta g h_1 W_A^3}{6\mu} \quad (9)$$

$$f_{2e} = \frac{\beta g W_A^2[2W_B^3 \Delta T - h_1(W_B^4 - W_A^4)]}{4\mu(W_A^3 + W_B^3)} \quad (10)$$

$$f_{3e} = \frac{6W_A^2 \sigma}{\rho B(W_A^3 + W_B^3)} \quad (11)$$

$$g_{1e} = \frac{\beta g h_1 W_B^3}{6\mu} \quad (12)$$

$$g_{2e} = \frac{\beta g W_B^2[2W_A^3 \Delta T + h_1(W_B^4 - W_A^4)]}{4\mu(W_A^3 + W_B^3)} \quad (13)$$

$$g_{3e} = \frac{6W_B^2 \sigma}{\rho B(W_A^3 + W_B^3)} \quad (14)$$

The terms on the righthand side of Eqs. 7 and 8 indeed represent the velocities caused by natural convection, additional convective strength, and forced convection, respectively. During the derivation of the above equations, all the physical properties were assumed constant and could be evaluated at the reference temperature. For the velocity distributions in the stripping section, Eqs. 6–14 are still valid except that the mass flow rate σ is replaced by $-\sigma$ and the subscript e by s.

Degree of separation

Material balances for each region in the enriching section can be expressed as follows.

$$\frac{\partial^2 C_{Ae}}{\partial \eta_A^2} - \frac{\partial}{\partial \eta_A}\left[\frac{\alpha C_{Ae}(1 - C_{Ae})h_1 W_A}{\tilde{T}}\right] = \left(\frac{W_A^2 V_{Ae}}{LD}\right)\frac{\partial C_{Ae}}{\partial \zeta} \quad (15)$$

$$\frac{\partial^2 C_{Be}}{\partial \eta_B^2} + \frac{\partial}{\partial \eta_B}\left[\frac{\alpha C_{Be}(1 - C_{Be})h_1 W_B}{\tilde{T}}\right] = \left(\frac{W_B^2 V_{Be}}{LD}\right)\frac{\partial C_{Be}}{\partial \zeta} \quad (16)$$

The boundary conditions are:

$$\frac{-\partial C_{Ae}}{\partial \eta_A} + \frac{\alpha C_{Ae}(1 - C_{Ae})h_1 W_A}{\tilde{T}} = 0, \quad \text{at } \eta_A = 0 \quad (17)$$

$$\frac{\partial C_{Be}}{\partial \eta_B} + \frac{\alpha C_{Be}(1 - C_{Be})h_1 W_B}{\tilde{T}} = 0, \quad \text{at } \eta_B = 0 \quad (18)$$

$$\begin{aligned}\frac{-\partial C_{Ae}}{\partial \eta_A} + \frac{\alpha C_{Ae}(1 - C_{Ae})h_1 W_A}{\tilde{T}} &= \frac{W_A}{W_B}\left[\frac{\partial C_{Be}}{\partial \eta_B} + \frac{\alpha C_{Be}(1 - C_{Be})h_1 W_B}{\tilde{T}}\right] \\ &= \frac{W_A \epsilon}{\delta}\left[\frac{\alpha C(1 - C)h_2 \delta}{\tilde{T}} - C_{Be} + C_{Ae}\right],\end{aligned}$$

$$\text{at } \eta_A = \eta_B = 1 \quad (19) \cdot (20)$$

$$C_{Ae} = C_{Be} = C_t, \quad \text{at } \zeta = 1 \quad (21)$$

where α is the thermal diffusion constant. In obtaining the above equations, some assumptions were made:

1. Neglect ordinary diffusion in the vertical direction and bulk flow in the horizontal direction.
2. Concentration changes in the range of 0.3 to 0.7 only for the entire column, or the separation is small. Therefore, the following terms

$$\frac{\alpha C_{Ae}(1 - C_{Ae})}{\tilde{T}}, \quad \frac{\alpha C_{Be}(1 - C_{Be})}{\tilde{T}} \quad \text{and} \quad \frac{\alpha C(1 - C)}{\tilde{T}}$$

in Eqs. 15–20 may be regarded as constant and all equal to θ. Consequently, the second terms on the lefthand sides of Eqs. 15 and 16 will vanish.

3. Within the barrier, no bulk flow exists and the change of concentration is small. Thus a linear relationship of concentration is expressed in Eq. 20. Moreover, due to the internal reflux at the end, the boundary condition is shown in Eq. 21.

Separation of variables in the form

$$C_{Ae}(\eta_A, \zeta) = \theta h_1 W_A \eta_A + \sum_{m=0}^{\infty} S_{Ae,m} F_{Ae,m}(\eta_A) G_{e,m}(\zeta) \quad (22)$$

$$C_{Be}(\eta_B, \zeta) = -\theta h_1 W_B \eta_B + \sum_{m=0}^{\infty} S_{Be,m} F_{Be,m}(\eta_B) G_{e,m}(\zeta) + \theta(\Delta T) \quad (23)$$

applied to Eqs. 15–20 leads to

$$G_{e,m}(\zeta) = \exp\left[-\lambda_{e,m}(1 - \zeta)\right] \quad (24)$$

$$F''_{Ae,m}(\eta_A) - \left[\frac{\lambda_{e,m} W_A^2 V_{Ae}(\eta_A)}{LD}\right] F_{Ae,m}(\eta_A) = 0 \quad (25)$$

$$F''_{Be,m}(\eta_B) - \left[\frac{\lambda_{e,m} W_B^2 V_{Be}(\eta_B)}{LD}\right] F_{Be,m}(\eta_B) = 0 \quad (26)$$

$$F'_{Ae,m}(0) = 0 \quad (27)$$

$$F'_{Be,m}(0) = 0 \quad (28)$$

$$-S_{Ae,m}F'_{Ae,m}(1) = \left(\frac{W_A}{W_B}\right) S_{Be,m} F'_{Be,m}(1) = \left(\frac{W_A \epsilon}{\delta}\right)\left[S_{Ae,m}F_{Ae,m}(1) - S_{Be,m}F_{Be,m}(1)\right] \quad (29) \cdot (30)$$

where the primes on $F_{Ae,m}(\eta_A)$ and $F_{Be,m}(\eta_B)$ denote the differentiations with respect to η_A and η_B, respectively.

Without loss of generality, we may assume the eigenfunctions $F_{Ae,m}(\eta_A)$ and $F_{Be,m}(\eta_B)$ to be polynomials.

$$F_{Ae,m}(\eta_A) = \sum_{n=0}^{\infty} d_{mne}\eta_A^n, \quad \text{with } d_{m0e} = 1 \quad (31)$$

$$F_{Be,m}(\eta_B) = \sum_{n=0}^{\infty} e_{mne}\eta_B^n, \quad \text{with } e_{m0e} = 1 \quad (32)$$

Define the ratio of expansion coefficients associated with eigenvalue $\lambda_{e,m}$ by

$$q_{e,m} = \frac{S_{Ae,m}}{S_{Be,m}} \quad (33)$$

Therefore, Eqs. 29 and 30 can be rewritten as

$$q_{e,m} = \frac{W_A \epsilon F_{Be,m}(1)}{W_A \epsilon F_{Ae,m}(1) + \delta F'_{Ae,m}(1)} = \frac{-W_A F'_{Be,m}(1)}{W_B F'_{Ae,m}(1)} \quad (34)$$

Substituting Eqs. 31 and 32 into Eqs. 25 and 26, all the coefficients d_{mne} and e_{mne} may be in terms of eigenvalue $\lambda_{e,m}$ after using Eqs. 27 and 28, as referred to in Appendix (i). Therefore, it is easy to solve all eigenvalues from Eq. 34 by use of an iteration method coupled with a false position method. Once the eigenvalue $\lambda_{e,m}$ is found, one may calculate $q_{e,m}$ from Eq. 34. In the meantime, the eigenfunctions associated with this eigenvalue are also completely specified by Eqs. 31 and 32. Inspection of Eqs. 25 and 26 shows that they are special cases of the Sturm-Liouville problem for which the velocity distributions in both regions have changed sign over the interval in question. Hence, there may exist both positive and negative sets of real eigenvalues that have the limit points $+\infty$ and $-\infty$, respectively. It is easy to find the orthogonality condition as

$$W_B \int_0^1 \left[\frac{W_A^2 V_{Ae}(\eta_A)}{LD}\right] S_{Ae,m} S_{Ae,n} F_{Ae,m}(\eta_A) F_{Ae,n}(\eta_A) d\eta_A + W_A \int_0^1 \left[\frac{W_B^2 V_{Be}(\eta_B)}{LD}\right] S_{Be,m} S_{Be,n} \cdot F_{Be,m}(\eta_B) F_{Be,n}(\eta_B) d\eta_B = 0 \quad (35)$$

when $n \neq m$.

For the stripping section, Eqs. 15–35 are still valid except that the subscript e is replaced by s, and Eqs. 21 and 24 are replaced by Eqs. 36 and 37, respectively.

$$C_{As} = C_{Bs} = C_b, \quad \text{at } \zeta = -1 \quad (36)$$

$$G_{s,m} = \exp\left[\lambda_{s,m}(1 + \zeta)\right] \quad (37)$$

Following the same procedures as those in the enriching section, we may obtain all the eigenvalues $\lambda_{s,m}$ and associated constants such as $q_{s,m}$. Since the velocity distributions in both regions change sign too, both positive and negative eigenvalues must be retained. However, in order to make the solution convergent, the negative set of eigenvalues for $\lambda_{e,m}$ and the positive set of eigenvalues for $\lambda_{s,m}$ should be omitted, as shown in Appendix (i). Until now, C_t, C_b, $S_{Ae,m}$ (or $S_{Be,m}$) and $S_{As,m}$ (or $S_{Bs,m}$) are still undetermined.

Combining concentration distributions, say Eqs. 22 and 23, in the enriching section with boundary conditions, Eqs. 21 and 36, gives

$$C_t - \theta h_1 W_A \eta_A = \sum_{m=0}^{\infty} S_{Ae,m} F_{Ae,m}(\eta_A) \quad (38)$$

$$C_t + \theta h_1 W_B \eta_B - \theta(\Delta T) = \sum_{m=0}^{\infty} S_{Be,m} F_{Be,m}(\eta_B) \quad (39)$$

$$C_b - \theta h_1 W_A \eta_A = \sum_{m=0}^{\infty} S_{As,m} F_{As,m}(\eta_A) \quad (40)$$

$$C_b + \theta h_1 W_B \eta_B - \theta(\Delta T) = \sum_{m=0}^{\infty} S_{Bs,m} F_{Bs,m}(\eta_B) \quad (41)$$

From the last four equations and the orthogonality conditions, we may obtain the general expressions for the expansion coefficients in both sections. Accordingly, for the enriching section, we have

$$W_B \int_0^1 \left[C_t - \theta h_1 W_A \eta_A\right] \cdot \left[\frac{W_A^2 V_{Ae}(\eta_A)}{LD}\right] S_{Ae,m} F_{Ae,m}(\eta_A) d\eta_A + W_A \int_0^1 \left[C_t + \theta h_1 W_B \eta_B - \theta(\Delta T)\right] \cdot \left[\frac{W_B^2 V_{Be}(\eta_B)}{LD}\right] S_{Be,m} F_{Be,m}(\eta_B) d\eta_B$$
$$= W_B \int_0^1 S_{Ae,m}^2 \left[\frac{W_A^2 V_{Ae}(\eta_A)}{LD}\right] \cdot F_{Ae,m}^2(\eta_A) d\eta_A + W_A \int_0^1 S_{Be,m}^2 \cdot \left[\frac{W_B^2 V_{Be}(\eta_B)}{LD}\right] F_{Be,m}^2(\eta_B) d\eta_B \quad (42)$$

Moreover, as shown in Appendix (ii), the eigenvalues with values of zero may be omitted during the calculation of C_t and C_b. After taking standard simplifying techniques with the use of Eqs. 25–30, the expansion coefficient associated with $\lambda_{e,m}$ can be expressed as

$$\begin{aligned} S_{Be,m} = \{ & -\theta h_1 W_A q_{e,m}(F'_{Ae,m}(1) - F_{Ae,m}(1) + 1) + (W_A/W_B) \\ & \cdot [\theta h_1 W_B(F'_{Be,m}(1) - F_{Be,m}(1) + 1) - \theta(\Delta T)F'_{Be,m}(1)]\} \\ & /\{q^2_{e,m}\lambda_{e,m}[F_{Ae,m}(1)(\partial F'_{Ae,m}(1)/\partial\lambda_{e,m}) - F'_{Ae,m}(1) \\ & \cdot (\partial F_{Ae,m}(1)/\partial\lambda_{e,m})] + (W_A\lambda_{e,m}/W_B)[F_{Be,m}(1) \\ & \cdot (\partial F'_{Be,m}(1)/\partial\lambda_{e,m}) - F'_{Be,m}(1)(\partial F_{Be,m}(1)/\partial\lambda_{e,m})]\} \end{aligned} \tag{43}$$

For the expansion coefficients in the stripping section, Eq. 43 is still valid with the subscript e replaced by s. To one's surprise, the intergrals adhering to C_t will vanish during the derivation of Eq. 43. This is due to the fact that velocity distributions change sign. Accordingly, once all the eigenvalues are found the associated expansion coefficients can be calculated from Eqs. 33 and 43. Now, only C_t and C_b remain undetermined.

Define the difference of average concentrations for region (A) in the enriching section by making use of Eqs. 22, 24, 25, and 27.

$$\begin{aligned} & \frac{\int_0^1 V_{Ae}(\eta_A)[C_{Ae}(\eta_A, 1) - C_{Ae}(\eta_A, 0)]d\eta_A}{\int_0^1 V_{Ae}(\eta_A)d\eta_A} \\ & = \frac{DL \sum\limits_{\substack{m=0 \\ \lambda_{e,m}\neq 0}}^{\infty} \left[\frac{S_{Ae,m}F'_{Ae,m}(1)}{\lambda_{e,m}}\right][1 - \exp(-\lambda_{e,m})]}{W_A^2 \int_0^1 V_{Ae}(\eta_A)d\eta_A} \\ & = C_t - \frac{\int_0^1 V_{Ae}(\eta_A)C_{Ae}(\eta_A, 0)d\eta_A}{\int_0^1 V_{Ae}(\eta_A)d\eta_A} \end{aligned} \tag{44}$$

Following the same definitions for other regions gives

$$\begin{aligned} & C_t - \frac{\int_0^1 V_{Be}(\eta_B)C_{Be}(\eta_B, 0)d\eta_B}{\int_0^1 V_{Be}(\eta_B)d\eta_B} \\ & = \frac{DL \sum\limits_{\substack{m=0 \\ \lambda_{e,m}\neq 0}}^{\infty} \left[\frac{S_{Be,m}F'_{Be,m}(1)}{\lambda_{e,m}}\right][1 - \exp(-\lambda_{e,m})]}{W_B^2 \int_0^1 V_{Be}(\eta_B)d\eta_B} \end{aligned} \tag{45}$$

$$\begin{aligned} & \frac{\int_0^1 V_{As}(\eta_A)C_{As}(\eta_A, 0)d\eta_A}{\int_0^1 V_{As}(\eta_A)d\eta_A} - C_b \\ & = \frac{DL \sum\limits_{\substack{m=0 \\ \lambda_{s,m}\neq 0}}^{\infty} \left[\frac{S_{As,m}F'_{As,m}(1)}{\lambda_{s,m}}\right][\exp(\lambda_{s,m}) - 1]}{W_A^2 \int_0^1 V_{As}(\eta_A)d\eta_A} \end{aligned} \tag{46}$$

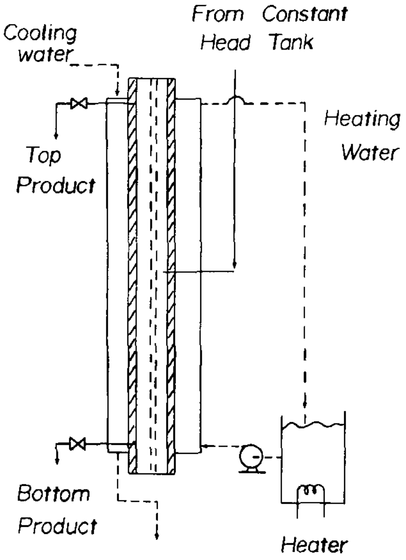

Figure 2. Flow diagram of a barrier thermal diffusion column.

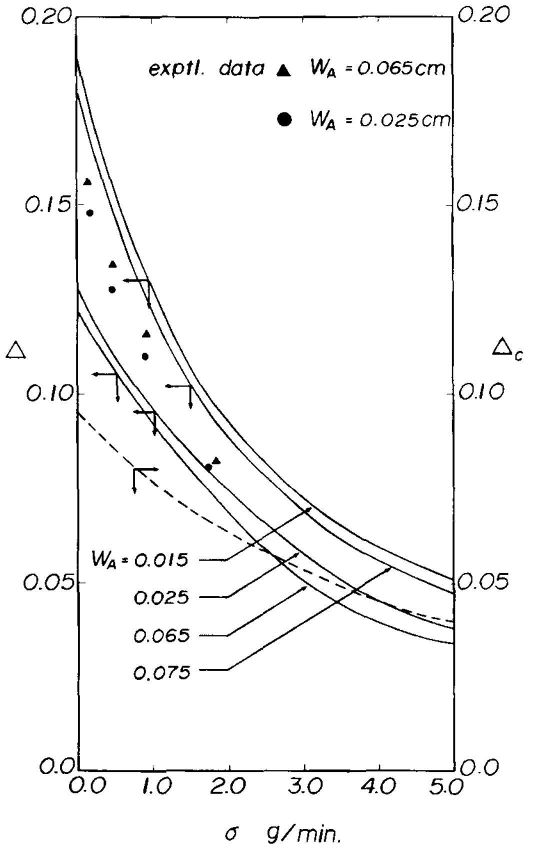

Figure 3. Benzene/*n*-heptane theoretical and experimental separations in improved column with theoretical separation in conventional tower for $W_A + W_B$ = 0.09 cm.

$$\frac{\int_0^1 V_{Bs}(\eta_B)C_{Bs}(\eta_B, 0)d\eta_B}{\int_0^1 V_{Bs}(\eta_B)d\eta_B} - C_b$$

$$= \frac{DL \sum\limits_{\substack{m=0 \\ \lambda_{s,m} \neq 0}}^{\infty} \left[\frac{S_{Bs,m}F'_{Bs,m}(1)}{\lambda_{s,m}}\right][\exp(\lambda_{s,m}) - 1]}{W_B^2 \int_0^1 V_{Bs}(\eta_B)d\eta_B} \tag{47}$$

Material balances around the feed positions lead to

$$\int_0^1 V_{Ae}(\eta_A)C_{Ae}(\eta_A, 0)d\eta_A - \int_0^1 V_{As}(\eta_A)C_{As}(\eta_A, 0)d\eta_A = C_f \int_0^1 [V_{Ae}(\eta_A) - V_{As}(\eta_A)]d\eta_A \tag{48}$$

$$\int_0^1 V_{Be}(\eta_B)C_{Be}(\eta_B, 0)d\eta_B - \int_0^1 V_{Bs}(\eta_B)C_{Bs}(\eta_B, 0)d\eta_B = C_f \int_0^1 [V_{Be}(\eta_B) - V_{Bs}(\eta_B)]d\eta_B \tag{49}$$

Combining Eqs. 44, 46, and 48 to eliminate the terms on the lefthand side of Eq. 48 yields an equation for C_t and C_b. After substituting the velocity distributions in the enriching section, one obtains

$$(3f_{1e} - 2f_{2e} + 2f_{3e})C_t - (3f_{1e} - 2f_{2e} - 2f_{3e})C_b$$

$$= 4f_{3e}C_f + \frac{12DL}{W_A^2}\left\{\sum_{\substack{m=0 \\ \lambda_{e,m} \neq 0}}^{\infty}\left[\frac{S_{Ae,m}F'_{Ae,m}(1)}{\lambda_{e,m}}\right][1 - \exp(-\lambda_{e,m})] + \sum_{\substack{m=0 \\ \lambda_{s,m0}}}^{\infty}\left[\frac{S_{Be,m}F'_{Be,m}(1)}{\lambda_{s,m}}\right][\exp(\lambda_{s,m})]\right\} \tag{50}$$

Similarly, from Eqs. 45, 47, and 49, we have

$$(-3g_{1e} + 2g_{2e} + 2g_{3e})C_t - (-3g_{1e} + 2g_{2e} - 2g_{3e})C_b$$

$$= 4g_{3e}C_f + \frac{12DL}{W_B^2}\left\{\sum_{\substack{m=0 \\ \lambda_{e,m} \neq 0}}^{\infty}\left[\frac{S_{Be,m}F'_{Be,m}(1)}{\lambda_{e,m}}\right] \cdot [1 - \exp(-\lambda_{e,m})] + \sum_{\substack{m=0 \\ \lambda_{s,m} \neq 0}}^{\infty}\left[\frac{S_{Bs,m}F'_{Bs,m}(1)}{\lambda_{s,m}}\right] \cdot [\exp(\lambda_{s,m}) - 1]\right\} \tag{51}$$

Once the concentration of products is calculated from Eqs. 50 and 51, the degree of separation for the whole column is found by

$$\Delta = C_t - C_b \tag{52}$$

Experiment

The experimental procedures are much the same as those in previous works (Cheuh and Yeh, 1967; Yeh and Ward 1971). Experiments were performed by employing a flat-plate thermal diffusion column with working dimension 123.8 × 10.8 cm, as shown in Figure 2. A stainless steel wire net of 0.02 cm thickness and 150 mesh was used as a barrier. The position of the barrier in the operating space was changed by placing Teflon sheets cut out as gaskets between the plates. The mean temperatures of the hot and cold walls were 322.5 and 280.5 K, respectively. The

Table 1. Eigenvalues and Related Expansion Coefficients for W_A = 0.04 and 0.05 cm

		W_A = 0.04 cm									
σ gpm	m	$\lambda_{e,m}$	$-\lambda_{S,m}$	$S_{Ae,m}$	$S_{Be,m}$	$S_{AS,m}$	$S_{BS,m}$	$F'_{Ae,m}(1)$	$F'_{Be,m}(1)$	$F'_{AS,m}(1)$	$F'_{BS,m}(1)$
0.15	0	0.0141	0.0143	1.5184	1.4928	−1.4873	−1.5129	-6.66×10^{-3}	8.47×10^{-3}	6.81×10^{-3}	-8.37×10^{-3}
	1	33.00	28.10	4.58×10^{-3}	-2.7×10^{-5}	6.2×10^{-5}	-5.25×10^{-3}	0.5432	114.54	48.77	0.7157
0.50	0	0.0464	0.0478	0.4664	0.4409	−0.4353	−0.4609	−0.0216	0.0286	0.0232	−0.0274
	1	33.42	28.84	4.58×10^{-3}	-2.5×10^{-5}	5.7×10^{-5}	-5.23×10^{-3}	0.5453	125.26	52.88	0.7185
1.50	0	0.1357	0.1488	0.1658	0.1403	−0.1344	−0.1608	−0.0607	0.1121	0.0934	−0.1220
	1	34.67	31.20	4.58×10^{-3}	-1.9×10^{-5}	4.5×10^{-5}	-5.19×10^{-3}	0.5513	162.10	67.62	0.7264
3.00	0	0.2636	0.3177	0.0907	0.0653	−0.0596	−0.0854	−0.1108	0.2405	0.1701	−0.1853
	1	36.73	35.58	4.58×10^{-3}	-1.3×10^{-5}	2.9×10^{-5}	-5.10×10^{-3}	0.5598	240.52	102.67	0.7379
5.00	0	0.4291	0.5937	0.0606	0.0353	−0.0295	−0.0556	−0.1655	0.3548	0.3445	−0.2856
	1	39.87	43.85	4.58×10^{-3}	-8×10^{-6}	1.5×10^{-5}	-4.95×10^{-3}	0.5705	415.99	204.27	0.7509
		W_A = 0.05 cm									
0.15	0	0.0148	0.0147	1.4267	1.4011	−1.4066	−1.4322	-8.84×10^{-3}	7.20×10^{-3}	8.96×10^{-3}	-7.04×10^{-3}
	1	26.60	31.24	5.25×10^{-3}	-6.1×10^{-5}	2.7×10^{-5}	-4.58×10^{-3}	0.7158	48.87	114.80	0.5432
0.50	0	0.0500	0.0485	0.4351	0.4094	−0.4150	−0.4405	-2.90×10^{-3}	0.0247	0.0304	−0.0229
	1	27.35	31.66	5.23×10^{-3}	-5.6×10^{-5}	2.5×10^{-5}	-4.58×10^{-3}	0.7187	53.26	126.25	0.5455
1.50	0	0.1561	0.1416	0.1518	0.1261	−0.1317	−0.1572	−0.0832	0.0802	0.0955	−0.0640
	1	29.74	32.92	5.18×10^{-3}	-4.4×10^{-5}	1.9×10^{-5}	-4.58×10^{-3}	0.7271	69.24	165.99	0.5518
3.00	0	0.3353	0.2748	0.0811	0.0553	−0.0610	−0.0864	−0.1561	0.1834	0.2059	−0.1163
	1	34.26	35.00	5.09×10^{-3}	-2.8×10^{-5}	1.3×10^{-5}	4.58×10^{-3}	0.7393	108.55	252.56	0.5608
5.00	0	0.6336	0.4477	0.0531	0.0269	−0.0328	−0.0580	−0.2396	0.3778	0.3825	−0.1728
	1	43.02	38.22	4.93×10^{-3}	-1.3×10^{-5}	7×10^{-6}	-4.58×10^{-3}	0.7523	230.96	453.42	0.5720

SI conversion: L/s = gpm × 0.0631.

plate temperatures were kept constant within ±0.5 K. Deaerated feed consisting of a 48.8 wt. % benzene and *n*-heptane mixture was introduced from a constant-head tank. Top and bottom products were withdrawn continuously at the same flow rate. Samples were analyzed at 30 min intervals with a refractometer until steady state was reached as indicated by changes of refractive index within ±0.0002. Some experimental data are shown in Figure 3.

The experimental results will be compared with theoretical predictions as follows. Some equipment parameters and physical properties of the mixture were found from the Perry *Handbook:*

$$\alpha = 1.2; \quad \epsilon = 0.378; \quad K = 3.8 \times 10^{-2}\ \mathrm{cal/cm \cdot S \cdot K}$$

$$K_0 = 3.2 \times 10^{-4}\ \mathrm{cal/cm \cdot S \cdot K}; \quad W_A + W_B = 0.09\ \mathrm{cm}$$

$$\mu = 1.34 \times 10^{-2} \exp(1{,}072/\tilde{T})\ \mathrm{cP}$$

$$\beta = 4.31 \times 10^{-2}/(550.5 - \tilde{T})^{0.641}\ \mathrm{g/cm^3 \cdot K}$$

$$\rho = 5.788(0.181 - 1.635 \times 10^{-4}\,\tilde{T})\ \mathrm{g/cm^3}$$

$$D = [2.47 + 4.65 \times 10^{-2}(\tilde{T} - 298)] \times 10^{-5}\ \mathrm{cm^2/S}$$

Two eigenvalues and related expansion coefficients for W_A = 0.04 and 0.05 cm have been calculated and are presented in Table 1 using σ as a parameter. However, only the first eigenvalue was found to be necessary during the evaluation of the degree of separation. The reason for this is understandable if one inspects the large value of the ratio of column length to column thickness in both sections. Some theoretical results, as well as those obtained in the Clusius-Dickel column with the distance between plates being ($W_A + W_B$), are also shown graphically in Figure 3 with σ as a parameter. Since the position of the barrier indeed has much influence on the performance, the first eigenvalues at various barrier positions are shown graphically in Figure 4 with mass flow rate as a parameter. Moreover, we also calculated the degree of separation at various barrier positions. Some results are shown in Figure 5 with σ as a parameter.

The improvement of separation achieved by operating at the optimal condition is best illustrated by calculating the percentage increase in separation based on the Clusius-Dickel column:

$$I = \frac{\Delta_{max} - \Delta_c}{\Delta_c} \tag{53}$$

Some results are presented in Table 2.

Summary

On the basis of this study, we conclude as follows.

The equation of separation in thermal diffusion columns with a permeable vertical barrier has been derived by using the orthogonal expansion technique. The eigenfunctions are expressed in terms of an extended power series expansion. The reasons we used the power series method with poor convergence are as follows:

1. Since the velocity distributions represented by Eqs. 7 and 8 involve η^3, the merit of expressing the eigenfunctions in terms of well-known functions, such as a confluent hypergeometric function, was lost.
2. Generally, the thermal diffusion column was designed with a large value of the ratio of column length to column thickness. Consequently, very few eigenvalues are necessary. In the extreme case, only the asymptotically lowest order eigenvalue will suffice.
3. It is more appropriate to use a power series to prove the convergence, as shown in Appendix (i).

In order to make the solution convergent, only the positive eigenvalues for the enriching section and the negative eigenvalues for the stripping section are necessary during calculation of the concentrations of products, as shown in Appendix (i). This behavior is quite different from that for the heat or mass transfer in countercurrent flow without recycle at the ends.

Two eigenvalues and associated expansion coefficients were calculated and are presented in Table 1 with the barrier position and mass flow rate as parameters. However, only the first eigenvalue was found to be necessary during the evaluation of the degree of separation.

The experimental data for the separation of a benzene and *n*-heptane mixture are plotted in Figure 3. Some theoretical results were also calculated and are presented in the figure, with the barrier position as a parameter. It is found from Figures 3 and 5 that the barrier position indeed has much influence on the

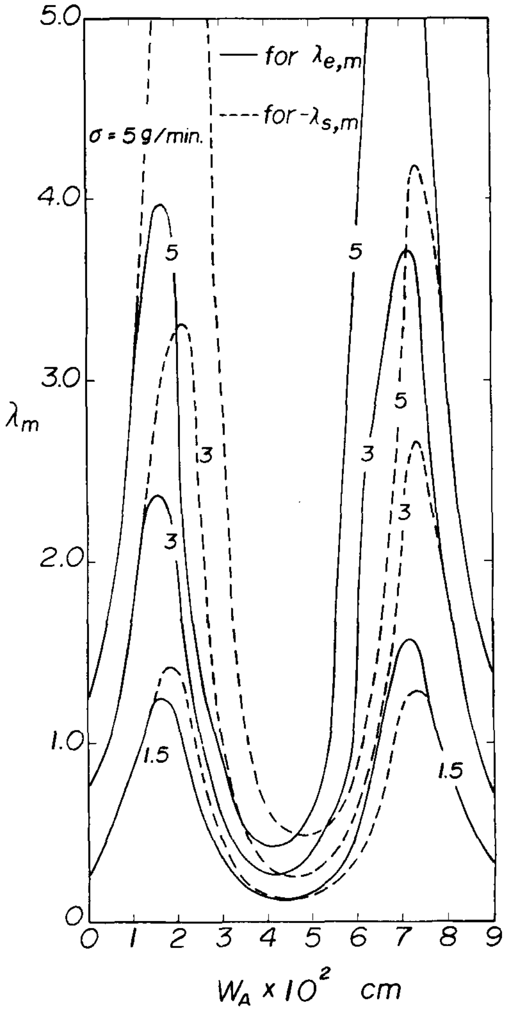

Figure 4. First eigenvalues for benzene/*n*-heptane mixture at various barrier positions with mass flow rate as parameter.

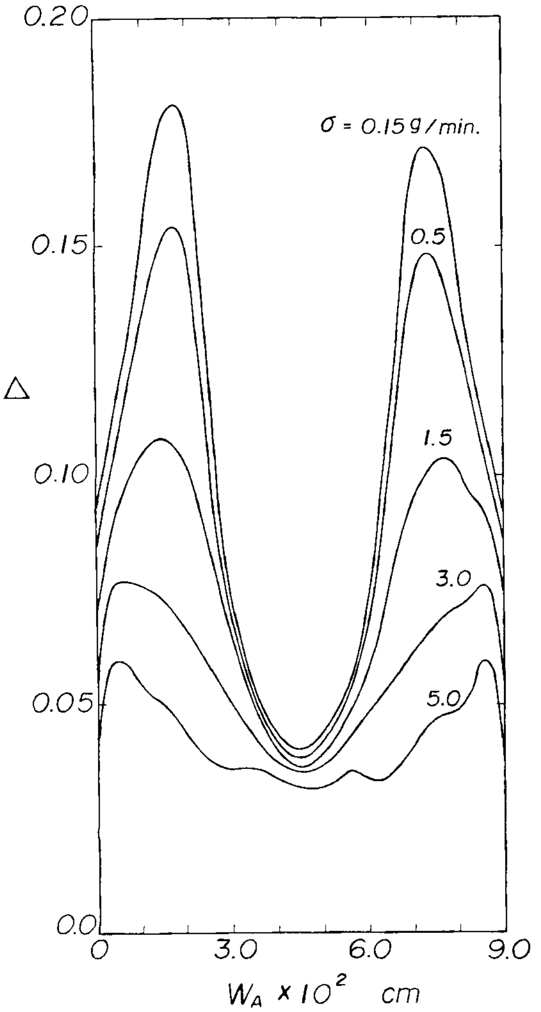

Figure 5. Effect of barrier position on degree of separation for benzene/*n*-heptane mixture at various flow rates.

Degree of separation for conventional column also shown at W_A = 0.0 or 0.09 cm.

degree of separation. Hence the experimental results with the degree of separation at $W_A = 0.065$ cm being larger than that at $W_A = 0.025$ cm, which is inverse to the theoretical results, may be due to the fact that the stainless steel net could not be placed at the exact position we wished. Moreover, a little departure of the experimental data from the theoretical results is due to the same reason. The degree of separation for the conventional column (Clusius-Dickel column) was also calculated and is presented in Figure 3.

Table 2. Comparison of Separation of Benzene/*n*-heptane Mixture Calculated for Clusius-Dickel Column and Barrier Column Operating at Best Barrier Position

σ	Δ_c	$W_{A,op}$	Δ_{max}	I
0.15	9.2	0.018	18.1	96.7
0.50	8.5	0.017	15.4	81.2
1.50	6.9	0.015	10.8	56.5
3.00	5.3	0.007	7.7	45.3
5.00	3.9	0.005	5.9	51.3

$C_f = 0.488$ for *n*-heptane.
SI conversion: L/s = gpm × 0.0631.

Since the position has much influence on the separation, the first eigenvalues at various barrier positions were calculated and are presented graphically in Figure 4. The degree of separation is also presented in Figure 5 with mass flow rate as a parameter. The degree of separation for the conventional column was calculated and is shown in the figure at $W_A = 0.0$ or 0.09 cm. It is found from Figure 5 that there exists an optimal position of the barrier for each σ. In general, the barrier should be located at the position near the hot or the cold wall in order to obtain maximum separation. This behavior is more evident as the mass flow rate increases. From the figure, it is also shown that minimum separation is reached when the barrier is located at the center.

Considerable improvement in separation is obtained when the position of the barrier is carefully chosen. Some numerical examples were calculated and are presented in Table 2.

During the evaluation of the separation, the most important assumption is $C(1 - C) \cong 0.25$ or $0.3 \le C \le 0.7$. When the concentration in the column is not in the range of $0.3 \le C \le 0.7$, the present analysis still applies if the degree of separation is not large. In this case, the nonlinear term $C(1 - C)$ may be approximated as $C_f(1 - C_f)$. For more intensive considerations of $C(1 - C)$, refer to Yeh and Chu (1975), Yeh and Chiou (1979), and Yeh and Yeh (1982).

It is stressed that the mathematical developments in this work may also be applied to other conjugated Graetz problems with recycle at both ends, which have not been mentioned yet in the literature.

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Notation

B = column width
C = weight fraction of component 1 in binary mixtures
C_b, C_f, C_t = C in bottom product, feed stream, and top product, respectively
D = ordinary diffusion coefficient in binary mixtures
d_{mn} = coefficient in the eigenfunction F_m for region (A)
e_{mn} = coefficient in the eigenfunction F_m for region (B)
F_m = eigenfunction associated with eigenvalue λ_m
f_1, f_2, f_3 = constants defined in the velocity distribution of region (A)
G_m = function defined during the use of orthogonal expansion method
g = gravitational acceleration
g_1, g_2, g_3 = constants defined in the velocity distribution of region (B)
h_1, h_2 = temperature gradient of the fluid in the slits and in the barrier, respectively
I = improvement of the degree of separation, Eq. 53
J_x = mass flux of component 1 of binary mixtures in the x direction
K, K_0 = thermal conductivity of the barrier and the fluid, respectively
L = one-half of the column length
P = pressure
q_m = ratio of expansion coefficients associated with eigenvalue λ_m
S_m = expansion coefficient associated with eigenvalue λ_m
$\bar{T}$ = reference temperature evaluated by Eq. 6
T_1, T_2 = mean temperature of the cold and the hot plates, respectively
V = velocity distribution of fluid in the vertical direction
W = thickness of the region
x = coordinate in the horizontal direction
z = coordinate in the vertical direction

Greek letters

α = thermal diffusion constant in binary mixtures
β = $-\partial\rho/\partial T$ evaluated at reference temperature
Δ = degree of separation defined by $C_t - C_b$
ΔT = $T_2 - T_1$
δ = thickness of the barrier
ϵ = permeability of the barrier
ζ = dimensionless coordinate in the vertical direction, defined by z/L
η = dimensionless coordinate in the horizontal direction, defined by x/W
θ = constant defined by $\alpha C(1 - C)/\tilde{T}$
λ_m = eigenvalue
μ = viscosity of the fluid
ρ = density of the fluid
σ = mass flow rate of top or bottom product

Subscripts

A = region between barrier and cold wall
B = region between barrier and hot wall
c = Clusius-Dickel columns
e = enriching section
s = stripping section
max = maximum quantity obtained at optimal conditions
op = optimal condition

Appendix

(i) Since all terms in Eqs. 44–47 have physical meaning, they must be finite. Now, consider the asymptotical value of the series on the righthand side of Eq. 44 as $\lambda_{e,m}$ approaches infinity. Let the velocity distributions in the enriching section be expressed as

$$V_{Ae}(\eta_A) = \sum_{i=1}^{3} a_i \eta_A^i \tag{A1}$$

$$V_{Be}(\eta_B) = \sum_{i=1}^{3} b_i \eta_B^i \tag{A2}$$

Substituting the above equations into Eqs. 25–28 and 31–32 yields

$$d_{m0e} = 1$$

$$d_{m1e} = 0$$

$$d_{m2e} = 0$$

$$d_{m3e} = \lambda_{e,m}/6, \propto \lambda_{e,m}$$

$$\vdots$$

$$\begin{aligned} d_{m(2n+1)e} = {} & \lambda_{e,m}(a_1 d_{m(2n-2)e} \\ & + a_2 d_{m(2n-3)e} + a_3 d_{m(2n-4)e}) \\ & /(4n^2 + 2n), \qquad \propto \lambda_{e,m}^n \end{aligned} \tag{A3}$$

$$e_{m0e} = 1$$

$$e_{m1e} = 0$$

$$\vdots$$

$$\begin{aligned} e_{m(2n+1)e} = {} & \lambda_{e,m}(b_1 e_{m(2n-n)e} \\ & + b_2 e_{m(2n-3)e} + b_3 e_{m(2n-4)e}) \\ & /(4n^2 + 2n), \qquad \propto \lambda_{e,m}^n \end{aligned} \tag{A4}$$

where $\propto$ means "proportional to." Therefore, retaining the highest order coefficients, say $d_{m(2n+1)e}$ and $e_{m(2n+1)e}$, we obtain from Eqs. 34, 43, and 44

$$q_{e,m} = \text{finite} \tag{A5}$$

$$\begin{aligned} \left[\frac{S_{Ae,m}F'_{Ae,m}(1)}{\lambda_{e,m}}\right][1 - \exp(-\lambda_{e,m})] = {} & \left[\frac{q_{e,m}S_{Be,m}F'_{Ae,m}(1)}{\lambda_{e,m}}\right] \\ & \cdot [1 - \exp(-\lambda_{e,m})], \quad \propto \frac{[1 - \exp(-\lambda_{e,m})]}{\lambda_{e,m}} \end{aligned} \tag{A6}$$

Obviously, if $\lambda_{e,m}$ approaches $-\infty$, divergence will occur. Following the same procedure for the stripping section gives

$$q_{s,m} = \text{finite}$$

$$\left[\frac{q_{s,m}S_{Bs,m}F'_{As,m}(1)}{\lambda_{s,m}}\right][\exp(\lambda_{s,m}) - 1], \quad \propto \frac{[\exp(\lambda_{s,m}) - 1]}{\lambda_{s,m}} \tag{A7}$$

Apparently, if $\lambda_{s,m}$ approaches ∞ divergence will occur. Consequently, the negative set of the eigenvalues for the enriching section and the positive set of the eigenvalues for the stripping section should be omitted.

(ii) Since $\lambda_{e,m} = 0 = \lambda_{s,m}$ are also eigenvalues, we first consider the case in the enriching section. As $\lambda_{e,m} = 0$, we obtain from Eqs. 25–32 that

$$F_{Ae,m} = F_{Be,m} = G_{e,m} = q_{e,m} = 1 \tag{A8}$$

Hence, Eq. 42 can be simplified as

$$\begin{aligned} S^*_{Be,m} = \Bigg(& C_t \left\{ \int_0^1 \left[\frac{W_A^2 V_{Ae}(\eta_A)}{LD}\right] d\eta_A \right. \\ & \left. + \frac{W_A}{W_B}\int_0^1 \left[\frac{W_A^2 V_{Be}(\eta_B)}{LD}\right] d\eta_B \right\} \\ & - \theta h_1 W_A \int_0^1 \left[\frac{W_A^2 V_{Ae}(\eta_A)}{LD}\right] \eta_A d\eta_A \\ & + \frac{W_A}{W_B}\int_0^1 [\theta h_1 W_B \eta_B - \theta(\Delta T)] \\ & \cdot \left[\frac{W_B^2 V_{Be}(\eta_B)}{LD}\right] d\eta_B \Bigg) \Bigg/ \left\{ \int_0^1 \left[\frac{W_A^2 V_{Ae}(\eta_A)}{LD}\right] d\eta_A \right. \\ & \left. + \frac{W_A}{W_B}\int_0^1 \left[\frac{W_B^2 V_{Be}(\eta_B)}{LD}\right] d\eta_B \right\} = S^*_{Ae,m} \end{aligned} \tag{A9}$$

Therefore, it results from Eqs. 22 and 23 that

$$C_{Ae}(\eta_A, \zeta) = \sum_{\substack{m=0 \\ \lambda_{e,m} \neq 0}}^{\infty} S_{Ae,m}F_{Ae,m}(\eta_A)G_{e,m}(\zeta) + \theta h_1 W_A \eta_A + S^*_{Ae,m} \tag{A10}$$

$$C_{Be}(\eta_B, \zeta) = \sum_{\substack{m=0 \\ \lambda_{s,m} \neq 0}}^{\infty} S_{Be,m}F_{Be,m}(\eta_B)G_{e,m}(\zeta) - \theta h_1 W_B \eta_B + \theta(\Delta T) + S^*_{Be,m} \tag{A11}$$

Obviously, the terms $S^*_{Ae,m}$ and $S^*_{Be,m}$ in Eqs. A10 and A11 have no influence on the determination of C_t, by inspection from Eqs. 44 and 45. Once C_t is solved, one may find the concentration at any position in the enriching section from Eqs. A10 and A11.

Following the same procedures for the stripping section, we obtain the same results as those in the enriching section. Consequently, it is only necessary to find the nonzero eigenvalues during the evaluation of concentrations of products at the ends.

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